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The overall purpose of this program has been to study the physics, technology, and spectroscopy of extreme ultraviolet (XUV) and soft x-ray lasers. The objective of the work was to develop a class of fixed frequency and tunable lasers whose wavelengths span the 100 A to 1000 A spectral region. Of special interest was developing incoherent, laser driven, harder x-ray sources that operate on a femtosecond time scale, and which could be used both for time resolved x-ray spectroscopy and, ultimately, for the pumping of solid state, micron dimension, x-ray lasers.

Work on XUV and soft x-ray lasers began about ten years ago with the use of high peak power microwaves to produce metastable, non-autoionizing core-excited species. The work has proceeded on both the spectroscopic and the laser fronts. Section 2 lists the principal accomplishments in these areas.

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Annual Technical Report

to

The Air Force Office of Scientific Research
&
The Army Research Office

for

Research Studies on Extreme Ultraviolet and Soft X-Ray Lasers

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For the Period
1 September 1988 — 31 August 1989

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Section 1

Introduction

The overall purpose of this program is to study the physics, technology, and spectroscopy of extreme ultraviolet (XUV) and soft x-ray lasers. The objective of our work is to develop a class of fixed frequency and tunable lasers whose wavelengths span the 100 Å to 1000 Å spectral region. We are also interested in developing incoherent, laser driven, harder x-ray sources that operate on a femtosecond time scale, and which could be used both for time resolved x-ray spectroscopy and, ultimately, for the pumping of solid state, micron dimension, x-ray lasers.

Our work on XUV and soft x-ray lasers began about ten years ago with the use of high peak power microwaves to produce metastable, non-autoionizing core-excited species. The work has proceeded on both the spectroscopic and the laser fronts. Section 2 lists the principal accomplishments in these areas, but we note that there have also been unexpected and especially fruitful interconnections between these areas: for example, our proposal for the super-Coster-Kronig Zn system, which initiated the successes in the 100 nm spectral region, both at Stanford and elswhere, to a large part resulted from the availability at Stanford of the Cowan atomic physics code. This code had been brought up to aid in our understanding of Column I and II quasi-metastable systems. Similarly, the successes of depletion spectroscopy have resulted from our use of laser produced x-ray excitation. This type of excitation, which is now key not only to our program, but also to programs at AT&T Bell Laboratories (Silvfast and Wood) and at Berkeley (Falcone), resulted from our attempts to produce larger densities of Li metastables, ionic and atomic, for store-and-transfer laser systems.

Other principal accomplishments of the laser portion of the program include the demonstration of large gain at 130.6 nm in the prototype super-Coster-Kronig Zn system of Mendelsohn and Harris, optimization of the Xe Auger laser first demonstrated by R. Falcone and his students at the Janus facility at Livermore, and the observation of gains of exp(80) at 96.9 nm in Cs.

The key accomplishment of the spectroscopic portion of our work has been the development and demonstration of a technique that allows a single visible laser to be used to define the core-excited manifold of atoms and ions. This technique also allows the measurement of autoionizing times which are sufficiently long that their Lorentzian width lies well under the combined Doppler–hyperfine structure.

New techniques for the amplification and compression of picosecond and femtosecond optical pulses have been developed recently. This technology, combined with the traveling-wave geometry we have developed, offers exciting new approaches for the development of lasers in the 100 Å–1000 Å spectral region.

We have made a number of advancements since we submitted our original proposal in March 1989, and these developments have suggested a few promising new projects that we would like to add to the program. We have submitted a supplemental proposal for this work. The major additional budget items represent improvements to our ultra-short pulse laser system: improved amplifier pump source, and oscillator stabilization. Other additions include studies in the new area of lasers without inversion, and development and optimization of the Xe III Auger 109 nm laser.

We note that the work described here has been, and will continue to be, jointly supported by other agencies, primarily the U. S. Office of Naval Research, and the U. S. Strategic Defensive Initiative Organization.

Section 2

Summary of Accomplishments

Significant accomplishments in the program supported by this contract or by predecessor awards are listed below in chronological order. Those marked with a star (⋆) have occured since our March 1989 proposal submission. Descriptions of our most recent work follows the list.

- Our work on metastable store-and-transfer began in 1975 with the proposal and first experimental demonstrations of laser-induced dipole–dipole, dipole–quadrupole and charge-transfer collisions.

- In 1977, the anti-Stokes radiation source was proposed and analyzed in the context of a two-photon blackbody.

- The anti-Stokes source was used to take the $3p$ absorption spectrum of neutral potassium at a resolution of $1.5\,\mathrm{cm}^{-1}$. This is the highest resolution ever obtained in this region of the spectrum.

- A class of quartet–doublet laser systems was proposed; the emission spectrum of neutral Li near 20 nm was taken, and the quartet and doublet manifolds of Li were experimentally connected for the first time.

- Microwave, and later, hollow-cathode technology, was used to produce metastable populations and to demonstrate several new types of core-excited spectroscopy. In one of these experiments the Grotrian diagram of core-excited Na was defined for the first time.

- The laser-produced plasma method of creating high densities of core-excited metastables was proposed and demonstrated.

- Using an impulsive electron source, produced by laser-produced x-rays, we demonstrated that levels which are imbedded in a continuum are not unusually sensitive to electron de-excitation.

- The ideas of shake-up and super-Coster-Kronig lasers were suggested.

- The concept of quasi-metastability was suggested and verified in several experiments.

- The method of core-excited depletion spectroscopy was suggested and used to define much of the $4p^5$ manifold of neutral Rb. It was found that the autoionizing times within this manifold are much longer than was first expected.

- Following the initial work of R. Falcone at Livermore, we observed and optimized gain for the Auger systems Xe ($\lambda = 108.9\,\mathrm{nm}$), Zn ($\lambda = 130.6\,\mathrm{nm}$), and Kr ($\lambda = 90.7\,\mathrm{nm}$).

- A new traveling-wave grazing incidence geometry was invented and used to construct a saturated 108.9 nm laser pumped with 3.5 J of 1064 nm radiation.

★ A saturated 96.9 nm laser in neutral Cs was constructed having an extrapolated small-signal gain of exp(83) in a total length of 17 cm.

★ Extension of the traveling-wave geometry to larger angles resulted in a 2 Hz Xe laser with gain of exp(33) pumped by only 0.5 J of 1064 nm radiation.

★ A careful parametric study of laser-produced plasmas showed it is possible to improve pumping efficiency for short pulses by more than a factor of 7 by prepulsing.

* A new theory was developed that shows that net laser gain is possible without a population inversion in certain cases of quantum mechanical interferences between levels.

Femtosecond Laser System

Oscillator Our efforts are presently centered on improving the stability of our femtosecond dye laser. This involves (1) pulse compression of the pump laser from 100 psec to 3 psec to provide stronger gain modulation of the dye laser and improve the synchronous mode-locking; (2) amplitude stabilization of the pump laser; (3) improved dye-jet nozzles made of sapphire; and (4) dye laser bandwidth control using a thick birefringent filter which has minimal etalon effects.

We have also tried to produce femtosecond pulses using additive-pulse mode-locking of the dye laser. However, no pulsewidth shortening was observed.

Pulsewidth expander/compressor We have improved the design of the pulse expander to maximize the power and bandwidth throughput of the pulse compressor, thereby minimizing truncation of the frequency spectrum which would result in temporal wings appearing on the pulse. The arrangement stretches 100 fsec-long pulses to 180 psec, allows 24 nm of bandwidth, and an output energy after the compressor of 50 mJ, limited by the compressor grating size.

Amplifier The amplification of the frequency chirped pulse occurs in Ti:sapphire, which provides a net gain of 10^{10}. Approximately 500 mJ of doubled Nd:YAG radiation is required to pump the Ti:sapphire crystal. Considerable effort was expended building a suitable pump laser from existing parts. Poor reliablity and insufficient 532 nm energy led us to purchase a commercial (Quantel YG682) Nd:YAG laser, which provides 800 mJ at 532 nm and acceptable beam quality.

The configuration of the Ti:sapphire amplifier has changed from an extensive multi-pass scheme to a simpler regenerative amplifier. Multi-pass designs require careful staging and isolation, while an unstable resonator regenerative amplifier can provide up to 100 mJ in a 200 psec–long pulse, starting from only a few tens of picojoules. Such an amplifier has been constructed and is currently being tested.

A single shot auto-correlator has been constructed, but not tested. This device will allow us to measure the pulsewidth of every high energy shot, using only the energy contained in a small reflection.

Lasers Without Inversion: Nonlinear Generation of VUV Radiation

It has previously not been possible to fully utilize the resonance properties of autoionizing levels for nonlinear generation of short-wavelength light due to the reabsorption of the created radiation. This greatly limits the interaction length and reduces the conversion efficiency of four-wave mixing. However, recent theoretical work in our group has shown that under the right circumstances a cancellation in the absorption of light can occur between two autoionizing lines. By utilizing this principle of absorption cancellation, nonlinear generation of short wavelengths may be greatly enhanced.

Two suitable autoionizing levels exist in zinc, with a cancellation wavelength of 104.8 nm. We propose to generate this radiation by four-wave mixing in zinc vapor. The conversion efficiency is predicted to increase by several orders of magnitude due to the presence of the absorption cancellation.

Section 3

Publications Supported

1. S. E. Harris, "Non-Reciprocity of Autoionizing Interferences: Lasers Without Inversion," *OSA Proceedings on Short Wavelength Coherent Radiation: Generation and Applications*, R. W. Falcone and J. Kirz, eds. (Optical Society of America, Washington, DC, 1988), Vol. 2, pp. 414–417.
2. C. P. J. Barty, D. A. King, G. Y. Yin, K. H. Hahn, J. E. Field, J. F. Young, and S. E. Harris, "12.8 eV Laser in Neutral Cesium," *OSA Proceedings on Short Wavelength Coherent Radiation: Generation and Applications*, R. W. Falcone and J. Kirz, eds. (Optical Society of America, Washington, DC, 1988), Vol. 2, pp. 13–20.
3. J. D. Kmetec, and S. E. Harris, "Targets for Efficient Femtosecond-Time-Scale X-Ray Generation," *OSA Proceedings on Short Wavelength Coherent Radiation: Generation and Applications*, R. W. Falcone and J. Kirz, eds. (Optical Society of America, Washington, DC, 1988), Vol. 2, pp. 209–212.
4. S. E. Harris and J. J. Macklin, "Lasers Without Inversion: Single Atom Transient Response," *Phys. Rev. A* **40**, 4135–4137 (October 1989).
5. A. Imamoğlu, "Interference of Radiatively Broadened Resonances," *Phys. Rev. A* **40**, 2835–2838 (September 1989).
6. S. E. Harris, A. Imamoğlu, and J. J. Macklin, "Nonreciprocal Emissive and Absorptive Processes," *Proceedings of NICOLS '89* (to be published).

Non-Reciprocity of Autoionizing Interferences: Lasers Without Inversion

S. E. Harris

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Abstract

Interferences of autoionizing lines may reduce or eliminate absorption of lower level atoms. Stimulated emission shows no such interferences, thereby allowing laser gain without population inversion.

Introduction

During the last several years we have made significant progress toward the realization of a class of extreme ultraviolet and soft x-ray lasers where the upper laser level is embedded in the continuum of the valence electron [1]. The advantage of this type of laser, as compared to more typical ionic lasers, is that the upper laser level occurs at much lower energy, and therefore may be pumped by cooler x-rays or electrons; this allows the operation of such lasers at pumping energies and powers which are much lower than would otherwise be possible. Highlights of recent work include: the demonstration that levels in the continuum may be pumped by electrons and retain their metastability under practical operating conditions [2]; the development of the technique of depletion spectroscopy for the measurement of autoionising lifetimes [3]: and most recently the demonstration of an extraordinary 12.8 eV laser in neutral Cs [4]. This laser achieves an equivalent small signal gain of exp(83), at a pumping energy of 3 joules in 20 psec.

An energy level diagram for the Cs laser is shown in Fig. 1. Though we have a good understanding of the mechanism for pumping the upper level, we do not understand how inversion is obtained; that is, why do electrons not populate the lower laser level? Though earlier proposals [1] suggested the use of a ionising laser to empty the lower level, no such laser was used in the Cs experiments.

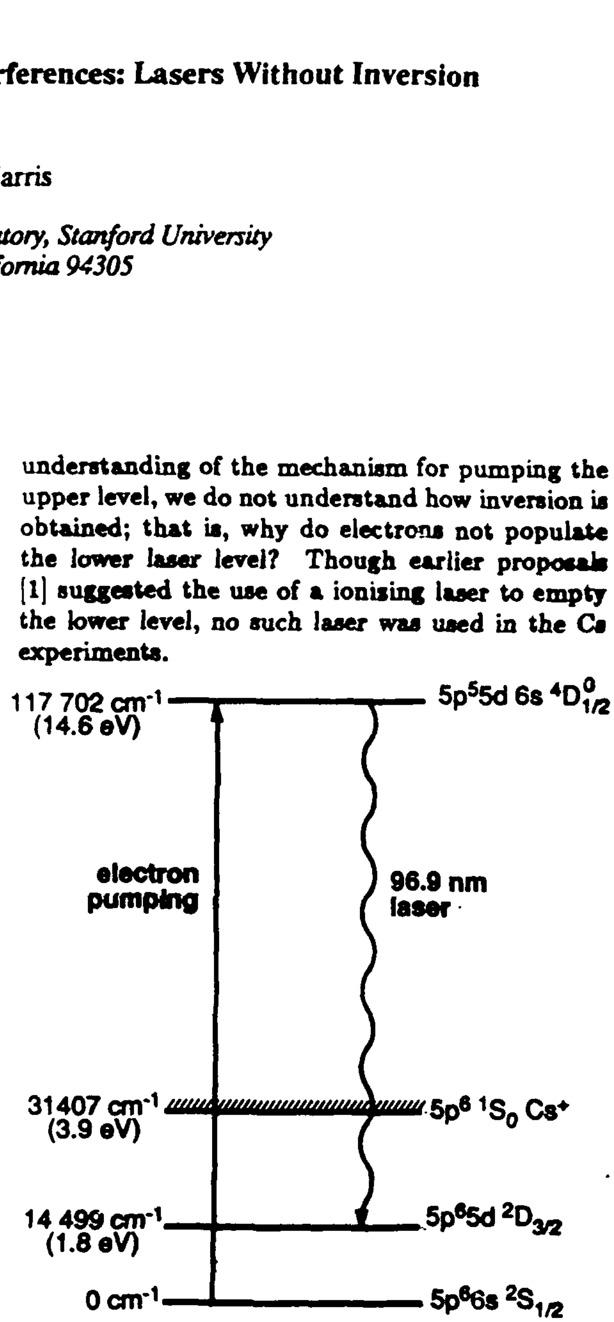

Figure 1—Partial energy level diagram for neutral Cs showing the laser transition.

In trying to understand this system the thought occured to me that perhaps inversion is not necessary. Perhaps, interference between two autoionizing levels could result in a cancellation of

12.8 eV Laser in Neutral Cesium

C. P. J. Barty, D. A. King, G. Y. Yin, K. H. Hahn, J. E. Field,
J. F. Young, and S. E. Harris

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Abstract

We report the operation of a saturated 12.8 eV (96.9 nm) laser in Cs vapor that has an extrapolated small signal gain of exp(83) in a total length of 17 cm. We believe that lasing occurs from a core-excited level embedded in the continuum of the valence electron. The laser is pumped by soft x-rays from a synchronous, traveling-wave, laser-produced (2.5 J, 15 ps, 1064 nm) plasma.

Introduction

Recently, it has been the aim of several research efforts to achieve lasing action at wavelengths below 100 nm. In general, successful efforts such as the 20.6 nm Se laser[1] have involved transitions in highly ionized atoms created within a dense laser-produced plasma and have required kilojoule class laser systems as pumping sources. In this work, we report the first operation of a new class of short wavelength lasers in which the upper level of the lasing transition is embedded in the continuum of the valence electron[2]. Consequently, it requires less than a joule of pumping energy to saturate the lasing transition.

We believe that the upper level of the laser is a core-excited level in neutral Cs and is pumped by photoelectrons generated by soft x-rays emitted from a laser produced plasma. Core-excited levels that are embedded within a continuum usually autoionize on a picosecond timescale, making the accumulation of population difficult. But this need not be the case; recent work by Spong *et al.*[3,4] has shown, for example, that there are many levels in neutral Rb that have autoionization lifetimes exceeding 10 ps and several that exceed 100 ps. Such long lifetimes can result either from angular momentum and spin selection rules that to first order prohibit autoionization, or from fortuitous radial matrix element cancellations. The possibility of using such levels to make extreme ultraviolet and soft x-ray lasers has been noted by several workers[5,6,7]. The existence of an inversion from an upper level embedded within a continuum has been inferred from fluorescence intensity measurements by Silfvast et al.[8].

Spectroscopy

An energy level diagram of the Cs 96.9 nm laser system is shown in fig. 1. The 117,702 cm^{-1} energy of the upper level has been measured by vacuum ultraviolet absorption spectroscopy [9,10], and the energy of the $5p^6 5d$ $^2D_{3/2}$ lower level is well known[10]. The difference, 96.897 nm, agrees with our measured emission wavelength of 96.86 ± 0.05 nm. The upper level designation is

Targets for Efficient Femtosecond-Time-Scale X-Ray Generation

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Abstract:

High power femtosecond lasers can be used to generate bright x-ray sources. The choice of element for the laser-produced-plasma target strongly affects the nature of the emitted x-rays. Combining low Z and high Z elements results in the brightest femtosecond x-ray radiators. Examples of lithium and tin combinations yielding 3.6 Å x-rays with up to several percent conversion efficiencies are given.

Introduction:

For many years, the output of pulsed laser systems have been focussed onto solid targets to generate plasmas. It is well known that a laser-produced- plasma can efficiently radiate in the XUV to x-ray regions of the spectrum. The recent development of very high peak power femtosecond lasers promises to yield even brighter plasmas emitting at even shorter wavelengths (1). We have studied the role of the laser target in a femtosecond-time-scale plasma and suggest several configurations which maximize the x-ray conversion efficiency while retaining the short pulse nature of the laser.

The dominant radiative processes in a laser-produced-plasma are line emission and two-body recombination. There have been experiments using picosecond and femtosecond lasers to drive x-ray-emitting plasmas (2-5), and also some measurements which show x-ray pulse lengthening well into the picosecond range (6). Both because of the relatively low power density on target, and because the targets were heavy metals, the electron temperatures were low, and the rise and fall times of the x-ray pulse were dominated by avalanche stripdown and two-body radiative recombination. Characteristic recombination times are slow compared to currently available femtosecond lasers. We propose conditions which maximize line radiation excited by hot electrons. The radiative rates and the heating and cooling times of the electrons can be much faster than recombination times.

The electrons are heated by the high power density of the laser, and cooled by inelastic ionizing collisions with the ions and by thermal diffusion. On a femtosecond time scale, ablation is negligible and thus very little cooling is achieved by plasma expansion. High Z elements have low thermal conductivity but very high inelastic cooling rates which prevent the laser from heating the electrons to high temperatures. Low Z elements quickly ionize completely and lose all inelastic

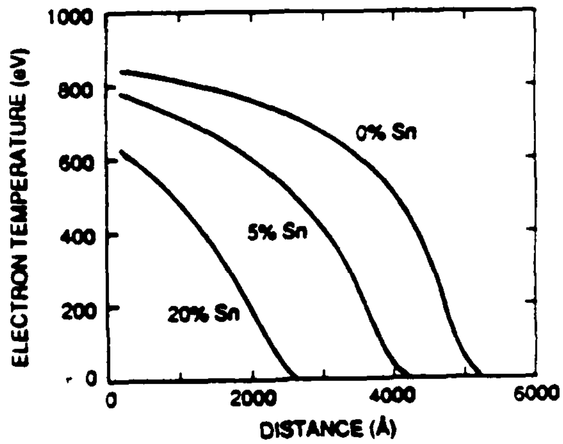

Figure 1: Electron temperature vs. distance for a solid Li-Sn mixed target at an absorbed energy of 1 kJ/cm^2, for different values of Sn density. The profiles are evaluated 50 fs after the peak of the laser pulse. Reproduced with permission from Ref. 7. Copyright 1988, American Physical Society.

Lasers without inversion: Single-atom transient response

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(Received 29 June 1989)

We discuss the effect of the transient response on the dynamics of lifetime broadened lasers that operate without the need for population inversion. A relationship between the steady-state absorptive transition probability rate and the transient gain and loss is given.

It has recently been shown that a population inversion is not a prerequisite for obtaining laser amplification and oscillation.[1-3] The essential idea is to utilize a system which causes a destructive interference in the absorption profile of lower-level atoms but not in the emission profile of upper-level atoms. An example of such a system is shown in the inset of Fig. 1(b). Level $|1\rangle$ is the lower laser level. Levels $|2\rangle$ and $|3\rangle$ are upper levels which are lifetime broadened with decay rates Γ_2 and Γ_3. This decay may result from autoionization, photoionization, tunneling, or spontaneous emission to a fourth level; but for the ideal case which we discuss here, the decay of both upper levels must occur to a common final continuum. (For example, for autoionization, this continuum is an ion and a free electron of prescribed angular momentum and arbitrary energy.)

In earlier work[1-3] the response of the lower-level atoms has been studied in the steady state, and the initial transient absorption caused by the excitation of the atoms at $t=0$ was ignored. Since this absorption effectively ends within several decay times of the upper levels, and since atoms may remain in level $|1\rangle$ indefinitely, the time-integrated contribution to the total absorption of this transient is vanishingly small, and is neglected when computing the steady-state (Fano-type) interference profiles of level $|1\rangle$ atoms. However, when operating at the zero of the interference profile, this transient absorption is the only absorption and should not be neglected. In fact, we show here, that this absorption determines what is, in effect, a threshold condition for lasers of this type.

When an atom is excited to level $|2\rangle$ or to level $|3\rangle$ (excitation occurs from some level or levels which are not shown), the stimulated response terminates within several decay times, and is, therefore, itself a transient response. For an ensemble of atoms, with rates into both upper and lower levels, the overall gain-loss balance is determined by the (single-atom) transient emission and the combined steady-state–transient absorption. It is often the case, for example in a cw discharge, that an ensemble of atoms is in steady state, while each individual atom has both a transient and a steady-state component. In this Rapid Communication we focus on the individual atom.

We use the equations of Ref. 1. These describe the threefold interference of level-$|1\rangle$ atoms to two upper levels and to the common continuum to which the upper levels decay. Results for a single level and a continuum, or two levels without a direct channel to the continuum, are obtained as special cases. The time-varying amplitudes of a lower level $|1\rangle$ and upper levels $|2\rangle$ and $|3\rangle$ are given by

$$\frac{\partial a_1}{\partial t}+j\Delta\tilde{\omega}_{11}a_1=\kappa_{12}a_2+\kappa_{13}a_3\,, \qquad (1a)$$

$$\frac{\partial a_2}{\partial t}+j\Delta\tilde{\omega}_{21}a_2=\kappa_{12}a_1+\kappa_{23}a_3\,, \qquad (1b)$$

$$\frac{\partial a_3}{\partial t}+j\Delta\tilde{\omega}_{31}a_3=\kappa_{13}a_1+\kappa_{23}a_2\,. \qquad (1c)$$

The quantities in these equations are

$$\begin{aligned}\Delta\tilde{\omega}_{11}&=-j\frac{W_C}{2},\quad \kappa_{12}=\tfrac{1}{2}[j\Omega_{12}+(\Gamma_2W_c)^{1/2}],\\ \Delta\tilde{\omega}_{21}&=\Delta\omega_{21}-j\frac{\Gamma_2}{2},\quad \kappa_{13}=\tfrac{1}{2}[j\Omega_{13}+(\Gamma_3W_c)^{1/2}]\,,\\ \Delta\tilde{\omega}_{31}&=\Delta\omega_{31}-j\frac{\Gamma_3}{2},\quad \kappa_{23}=-\tfrac{1}{2}(\Gamma_2\Gamma_3)^{1/2},\end{aligned} \qquad (2)$$

where Γ_2 and Γ_3 are the decay rates of levels $|2\rangle$ and $|3\rangle$; $\Delta\omega_{21}=\omega_2-(\omega_1+\omega)$, $\Delta\omega_{31}=\omega_3-(\omega_1+\omega)$, and ω is the angular frequency of the electromagnetic field; Ω_{12} and Ω_{13} are the respective Rabi frequencies ($\mu E/\hbar$), and W_c is the (direct channel) photoionization rate of level $|1\rangle$ to the continuum. We assume that the basis set has been prediagonalized[1] so that Γ_2, Γ_3, and W_c are real. We note the importance of the cross term κ_{23}, which represents the fact that as level $|2\rangle$ decays it drives level $|3\rangle$ and vice versa. This term arises since both levels couple to the same continuum level and therefore to each other.

We begin by examining computer solutions of these equations. The parameters for the computer runs (Fig. 1) are chosen so as to attain a zero in the steady-state absorption. The parameters for the three-level system are $\Omega_{12}=1/\sqrt{10}$, $\Omega_{13}=1$, $\Delta\omega_{21}=-5$, $\Delta\omega_{31}=50$, $\Gamma_2=1$, $\Gamma_3=10$, and $W_c=0$. For comparison, we also show a two-level system with the same parameters but without the interfering level $|3\rangle$.

Figure 1(a) shows the probability for level $|1\rangle$ occupancy, $|a_1(t)|^2$, versus time for the two-level system with the boundary condition $a_1(0)=1$, $a_2(0)=a_3(0)=0$. The absorption consists of the sum of a transient term and of a (golden-rule) steady-state term. Figure 1(b) shows this same quantity for the ideal three-level system. Here the steady-state term is zero (zero slope) and the absorption consists of only the transient term.

Figures 1(c) and 1(d) show the emission process. Here the boundary condition at $t=0$ is $a_2(0)=1$, $a_1(0)$

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Interference of radiatively broadened resonances

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(Received 31 May 1989)

We show that a three-state atomic system where the upper two states have the same J and m_J quantum numbers and decay radiatively to the states of a single atomic level is equivalent to the recently proposed inversionless laser system [S. E. Harris, Phys. Rev. Lett. **62**, 1033 (1989)].

In a recent paper, Harris[1] showed that in some atomic systems population inversion is not a necessary condition for obtaining laser action. He considered a three-level system, where the two upper levels are purely lifetime broadened and decay by autoionization to an identical continuum. For this system, stimulated emission and absorption line shapes are different due to the presence of Fano-type interferences.[2,3] At a certain laser frequency, the absorption rate goes to zero, whereas the emission rate remains nonzero. Amplification of a laser field at this frequency is possible even though the number of lower-level atoms in the system is higher than the number of upper-level atoms. A semiclassical approach was used in the analysis of Ref. 1, thus radiatively broadened resonances were not considered.

In this Rapid Communication, we use a full quantum-mechanical approach to show that a three-state atomic system where the upper two states are radiatively broadened is equivalent to the lifetime broadened systems considered by Harris, provided certain selection rules are satisfied. The selection rules require that the two upper states have the same J and m_J quantum numbers and that they decay to the states of a single atomic level.

The system that we consider is outlined in Fig. 1. A lower atomic state $|1\rangle$ is coupled to two upper states $|2\rangle$ and $|3\rangle$ via a probe laser field at frequency $\omega_{\mathbf{k}_L}$. These upper states decay to a single atomic state $|i\rangle$ by spontaneous emission. The emitted photon can have arbitrary direction and energy.[4] For absorption, we consider an atom which has been in the lower state for a long time compared to the lifetimes of the upper states. In the absence of dephasing events, the interaction of such an atom with a weak monochromatic incident field leads only to Raman scattering into state $|i\rangle$. Raman scattering for this system takes place through two intermediate states ($|2\rangle$ and $|3\rangle$), so that the resulting scattering (identically equal to absorption) probability has an interference term. This interference term may yield a zero in the absorption line shape.

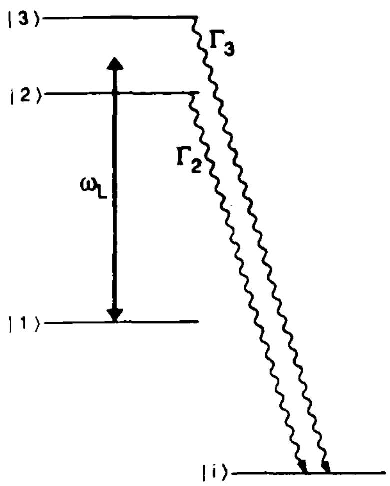

FIG. 1. Radiatively broadened system.

For emission, we consider an atom pumped into state $|2\rangle$ from a reservoir. Due to the coupling of the states $|2\rangle$ and $|3\rangle$ via their decay process, state $|3\rangle$ is also excited, and therefore the stimulated transition to state $|1\rangle$ takes place through two paths. The corresponding interference term, however, is different in this case, giving an emission line shape that is not the same as the absorption.

The basis set that we use in the analysis consists of a number of eigenstates of the noninteracting atom plus radiation field Hamiltonian. Assuming that there are no photons in any but the laser mode of the radiation field initially, we can write the state vector of the total system in the interaction representation[5,6] as

$$|\Psi_I(t)\rangle = a_1(t)|1,n_{\mathbf{k}_L}\rangle + a_2(t)|2,n_{\mathbf{k}_L}-1\rangle + a_3(t)|3,n_{\mathbf{k}_L}-1\rangle + \sum_{\sigma\mathbf{k}} a_{i,\sigma\mathbf{k}}(t)|i,n_{\mathbf{k}_L}-1,1_{\sigma\mathbf{k}}\rangle , \tag{1}$$

where a_1, a_2, a_3, and $a_{i,\sigma\mathbf{k}}$ are the probability amplitudes; $n_{\mathbf{k}_L}$ is the number of photons in the laser mode; $1_{\sigma\mathbf{k}}$ represents the fact that the number of photons in the radiation mode $\sigma\mathbf{k}$ is one; $\mathbf{k}$ and σ are the wave vector and polarization of the spontaneously emitted (or Raman scattered) photon, respectively. The expansion in (1) assumes that the radiative decay into atomic state $|1\rangle$ is negligible and that only the eigenstates with approximately equal energy are coupled by the interactions. The latter assumption is equivalent to the rotating wave approximation in the semiclassical approach.

By substituting (1) in Schrödinger's equation[6]

$$i\hbar \frac{\partial |\Psi_I(t)\rangle}{\partial t} = \hat{H}_I(t)|\Psi_I(t)\rangle , \tag{2a}$$

where

Nonreciprocal Emissive and Absorptive Processes

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1. Introduction

Though it has long been believed that an inversion is critical to obtaining laser amplification and oscillation, this is not the case. Recently, we have shown that if two levels decay to an identical continuum, that this decay couples these levels and results in nonreciprocal emissive and absorptive profiles (1,2). Though previous work (1-3) has emphasized systems where the decay results from autoionization or photoionization, these ideas also hold for purely radiative decay (4). Figure 1 shows such a system. Two upper states of the same angular momentum radiatively decay to the same final level and are coupled through this decay. In a sense, one may look at this coupling as an "internal radiative trapping." When a photon is spontaneously emitted by an upper state, it has a probability of being instantaneously reabsorbed by the other upper state of the same atom. It is in this sense that the upper states are coupled.

When one examines the absorption profile of atoms which are in state |1>, one finds interferences in the absorption profile - which in this case can be viewed as interferences in the anti-Stokes scattering profile. These interferences are not present in the emissive profile of atoms which at t=0 are in state |2> or |3>.

2. Non-Cancelable Channels

In previous work, we have studied the ideal case, where both upper levels decay to the same final state. There are always other non-cancelable channels which we have neglected. In particular there is always spontaneous emission on the laser channel itself. The assumption is therefore that the decay rate of the cancelable channel is much larger than the decay rate of the non-cancelable channels. For this condition, it may be shown that the ratio of the stimulated emission cross section to the absorption cross section is equal to the ratio of the cancelable and non-cancelable decay rates.

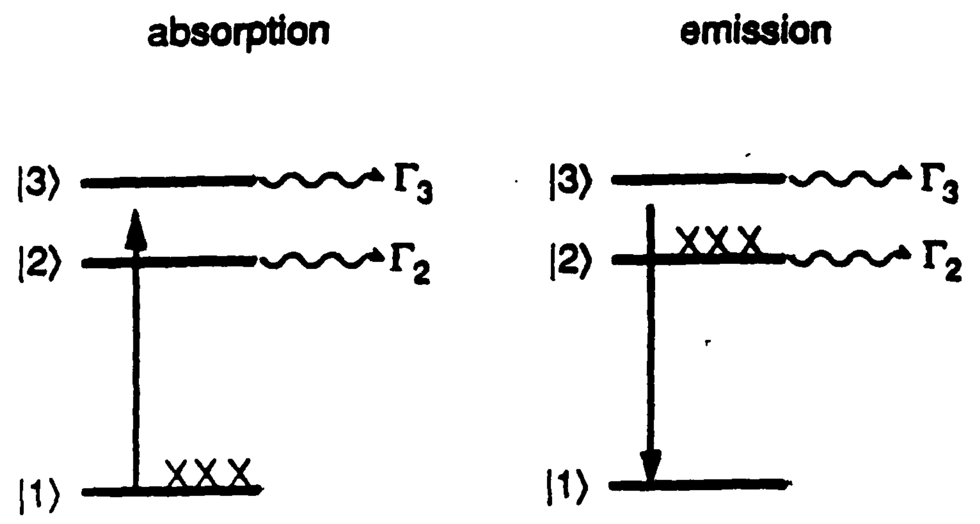

Fig. 1 Schematic of a system where the interference results from radiation broadening. The selection rule for this type of sytem is: states |2> and |3> must have the same parity, J, and m_J; and must decay to a common final level (not shown in the figure).